FLARE R&D FOR ULTRA PURE LIQUID ARGON

B. J. Hansen, M. J. White

Fermi National Accelerator Laboratory
Batavia, Illinois, 60510, USA

ABSTRACT

FLARE (Fermilab Liquid ARgon Experiments) is an initiative to construct several liquid argon detectors to further exploit the two Fermilab neutrino beams, the operational Booster beam, and the recently completed NuMI beam. For optimum physics results, a liquid argon impurity level of less then 0.1 ppb oxygen equivalent is required. The necessary R&D program has been established and the experimental setup for the first stage is nearing completion. The purpose of the first stage is to gain experience in achieving, maintaining, and monitoring ultra pure liquid argon levels. An overview of the technology and methods utilized in the FLARE R&D program are discussed.

INTRODUCTION

Fermilab's recent major investments in neutrino physics, the construction of two intense neutrino beams spanning energies from 0.5 GeV to 20 GeV, opens many doors to a variety of new neutrino experiments. The goal of FLARE is to further utilize the potential of Fermilab's neutrino beams, with a series of experiments [1]:

- A large, 15 kton class, LAr detector in the off-axis NuMI beam.
- A small, 40 ton class, LAr detector for the Booster beam or the near NuMI hall.
- Another dedicated LAr detector to search for neutrinoless double beta decay.

All of these use the recently developed detector technology of the Liquid Argon Time Projection Chamber (LAr TPC). ICARUS has already demonstrated the technical aspects for the success of a 500 ton class LAr detector with their construction and operation of the

ICARUS T600 detector [2]. The FLARE R&D program is therefore acting as a technology transfer to further establish credibility of technical claims and cost estimates.

The operating principle of the LAr TPC is based on the fact that in highly purified LAr, free electrons produced by the ionization of argon atoms can be transported practically undistorted by a uniform electric field over distances of several meters [2]. This translates to the necessity of reducing the presence of any electro-negative impurities to extraordinary levels, on the order of 0.1 ppb oxygen equivalent. Therefore, industrial argon with an initial impurity level of 1-2 ppm oxygen must be purified during the filling stage and then preserved throughout the entire lifetime of the experiment. In addition, the level of impurities must be accurately monitored during the various phases of the detector operation to insure consistent performance.

SYSTEM OVERVIEW

A simplified schematic of the experimental setup is presented in FIGURE 1. The cryogenic system consists of a vacuum insulated 170 liter dewar utilized as a housing chamber for the detector (purity monitor). In addition, a 120 liter liquid helium quality dewar is utilized as a means for storing and supplying liquid argon. The purifier setup is vacuum insulated and is installed between the detector chamber and storage dewar via vacuum insulated transfer line. Two identical condensers, one for the detector chamber and one for the storage dewar, are used to carry out pumpless transfers for the purification process, as well as to retain the liquid argon within the system. Liquid level probes are utilized to prevent overfilling and allow the flexibility to make transfers of various volumes.

The purification process is accomplished by simply cycling the liquid argon back and forth between the detector chamber and the storage dewar, where by means of a check valve, the argon is flushed through the purifiers only during transfers to the detector chamber. The transfers are carried out by establishing a pressure differential between the two dewars.
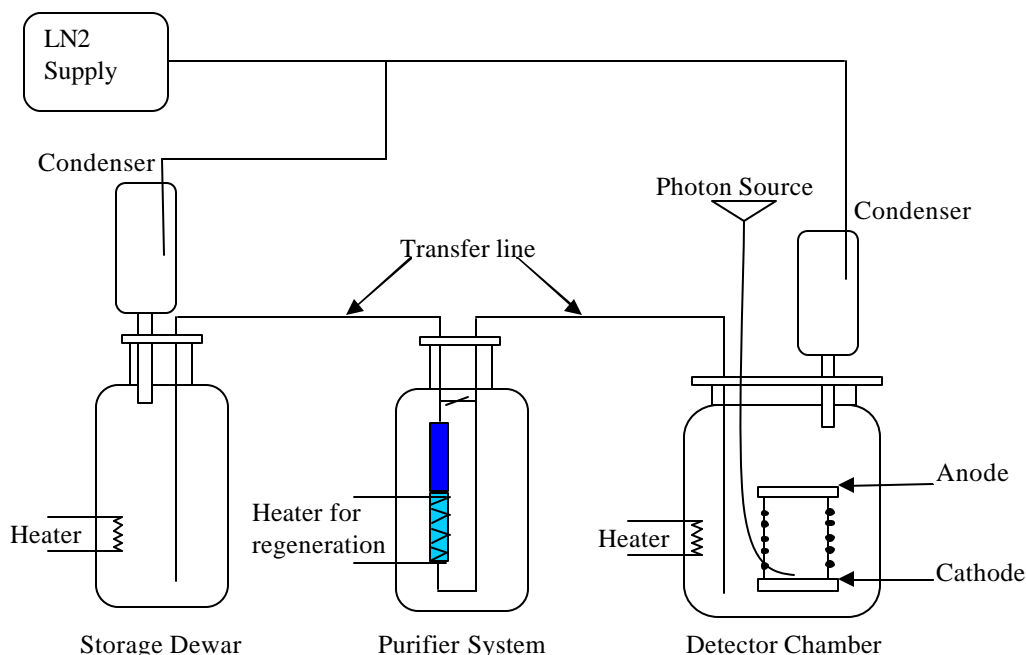

FIGURE 1. Simplified schematic of experimental system.

The high pressure of the supply dewar is initiated by decreasing its condenser output and activating a 100 W heater, while the low pressure of the receiving dewar is initiated by increasing its corresponding condenser output. Once the high and low set pressures have been established, they are stabilized and maintained by adjusting the inflow of liquid nitrogen into the condensers.

One full volume purification cycle will take approximately 6-8 hours. Between each cycle the purity within the detector dewar can be determined by using the electron lifetime purity monitor described in a later section. Once the desired purity is achieved, it can be maintained by establishing a recirculation process of smaller volume transfers. The rate and size of recirculation to maintain a desired purity will be a function of the contamination into the system and effectiveness of the purifiers.

During initial transfers there is an increase in demand put on the condenser of the receiving dewar, due to the rapid boil off of liquid argon in contact with the warm surfaces of the transfer line and receiving dewar inner walls. Once the purification process is completed the pressure of the system is stabilized to a constant operating pressure and the condensing demand is then limited to the steady state heat leak into the system.

CONDENSER DESIGN

The large range of condensing demands, from initial transfers to steady-state boil off, has resulted in a hybrid design of vertical tapered copper tubing inside a liquid nitrogen bath. The condenser design in FIGURE 2 is presented horizontally but operates in the vertical position. The condensers are installed vertically on top of the detector chamber and storage dewar as seen in FIGURE 1. As liquid argon is boiled off, the innermost stainless steel tube transfers the argon vapor to the top of the condenser and into the tapered copper tubing. On the opposite side of the copper tubing is the liquid nitrogen bath, which provides the source for cooling. The argon condenses along the surface corresponding to the liquid nitrogen fill height and drains down into the dewar. The separation of the upward flow of vapor from the downward drainage of condensate eliminates vapor shear from acting on the surface of the film condensate, which at high vapor flow rates (during initial transfers), could cause the condenser to drain improperly and flood.

The two-tier condenser boasts a high heat transfer capacity and provides ease of control at both low and high condensing demands. Where as, if the condenser utilized only a single large diameter copper tube at low condensing demands, the nitrogen liquid level needed to maintain pressure would be extremely low. Furthermore, a slight rise in nitrogen liquid level would result in a large rise in the effective heat transfer area, making it difficult to maintain a steady pressure.
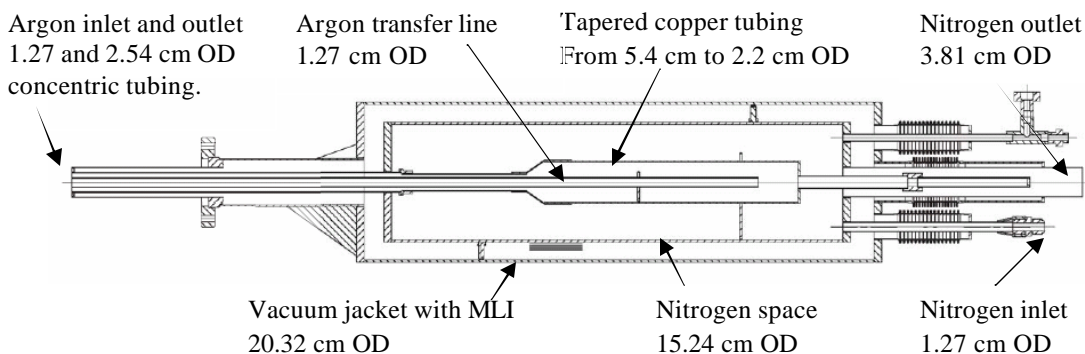.

Argon inlet and outlet 1.27 and 2.54 cm OD concentric tubing.

Argon transfer line 1.27 cm OD

Tapered copper tubing From 5.4 cm to 2.2 cm OD

Nitrogen outlet 3.81 cm OD

Vacuum jacket with MLI 20.32 cm OD

Nitrogen space 15.24 cm OD

Nitrogen inlet 1.27 cm OD

FIGURE 2. Condenser design with an overall length of 1.3 meters.

Therefore, at low condensing demands it would be more difficult to fine tune the controller for a large single diameter condenser then for the smaller diameter of the two-tier condenser.

A simple heat transfer model was created to aid in the development of the condenser design and then to predict the theoretical performance of the final design. The model describes the heat transfer process from the condensing argon vapor through the frozen film of argon and copper tubing, to the boiling nitrogen bath.

Theoretical Condenser Performance

Initially, cool down begins due to heat transfer from the warm vapor to the cold inner heat transfer surface. When the temperature of the vapor is reduced to its saturation temperature and the latent heat of the vapor is removed, condensate is formed. As a result, a liquid film develops along the cold surface and gravity drains the condensate downward. Condensation now occurs between the liquid-vapor interface and transfers heat by conduction through the liquid film to the cold surface. The liquid film thickness continues to increase as it drains downward and acts as a resistance to heat transfer between the vapor and the cold surface. Therefore, an increase in nitrogen liquid level results in an increase in average liquid film thickness and thereby decreases the heat transfer effectiveness.

The intended operating temperature of liquid nitrogen at atmospheric pressure (77.4 Kelvin) causes the temperature of the copper surface to drop below the melting point of argon (83.6 Kelvin). Consequently, a layer of frozen argon exists between the liquid film condensate and the copper surface. The film thickness of frozen argon was found to be much less than one millimeter. The relatively thin film of frozen argon is attributed to its low coefficient of thermal conductivity (0.2-0.4 W/m*K), in addition to the small temperature gradient (1-2 Kelvin) between the below-freezing copper surface and the above-freezing argon solid-liquid interface [5].

Some data produced from the heat transfer model is presented in FIGURES 3 and 4. FIGURE 3 is a graph of nitrogen liquid level along the first tier of the condenser versus heat transfer at various argon pressures. The insulating effect of larger liquid argon film thicknesses, corresponding to larger nitrogen liquid levels, is noted by the nonlinear increase in heat transfer with nitrogen liquid level. From FIGURE 4 it is noted that the maximum overall heat transfer capacity ranges from 500-1500 W for corresponding argon pressures ranging from 100-250 kPa.
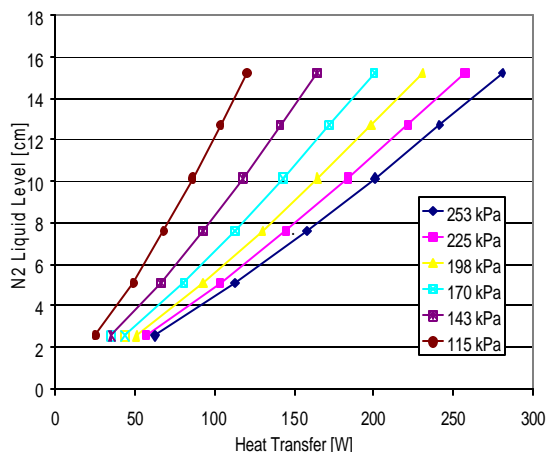
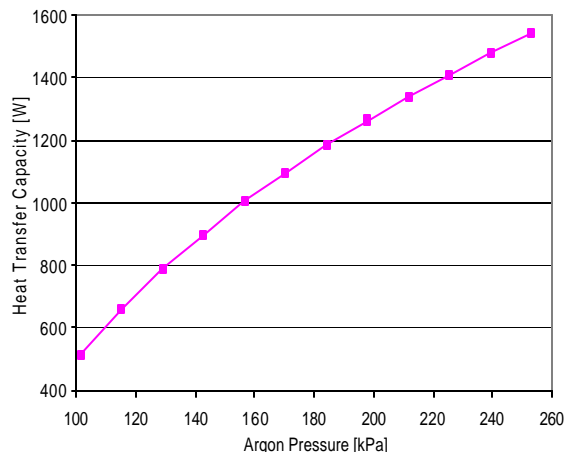**FIGURE 3.** Lower tier heat transfer results.   **FIGURE 4.** Condenser heat transfer capacity.

PURIFICATION AND REGENERATION

There are two major goals of FLARE'S purification R&D. The foremost goal is to purify the liquid argon of electronegative impurities to less than 0.1 ppb of oxygen equivalent. This level of purity is required to produce the necessary free electron lifetimes for physics experiments. The secondary goal is to maintain the desired purity level of 0.1 ppb for extended periods of time in an economical manner. Traditionally fluids are purified by an adsorber, which must be sent back to the manufacturer for regeneration once it has been saturated with oxygen. Taking the adsorber out for regeneration allows a small amount of air to enter the system during the adsorber exchange. This small amount of air will significantly decrease the purity of the argon and cause the adsorber to saturate quickly. So a series of experiments were conducted to see if an oxygen adsorber could be regenerated in-line, thus saving time, money, and purity level.

The adsorber test setup shown in FIGURE 5 is designed to test in-line regeneration, to determine the quantity of oxygen adsorbed, and to determine the purity of argon leaving the adsorber. The basic procedure for regeneration is as follows. The entire system is evacuated to remove all oxygen from the system. The oven is used to preheat the adsorber to 250 °C. Next a mixture of 5% hydrogen and 95% argon by volume content is opened and gas is allowed to flow through a gas preheater, the heated adsorber, and then finally vented to the atmosphere. A preheater is used so that gas coming into the adsorber is not significantly cooling down the adsorber pellets, thus allowing for a more even temperature distribution.

The hydrogen reacts exothermically with the metal oxides in the adsorber to produce water vapor and results in a further increase in adsorbent temperature. The pellets in the adsorber can start to break down above 300 °C. Therefore, temperature sensors were mounted both inside the adsorber and the oven to monitor the adsorber temperature throughout the process. The heater was turned off periodically to maintain a temperature around 250 °C. Regeneration takes approximately an hour, after which the adsorber is allowed to cool and the system is evacuated. The adsorber is then ready for testing in-place or removed for use in another system.
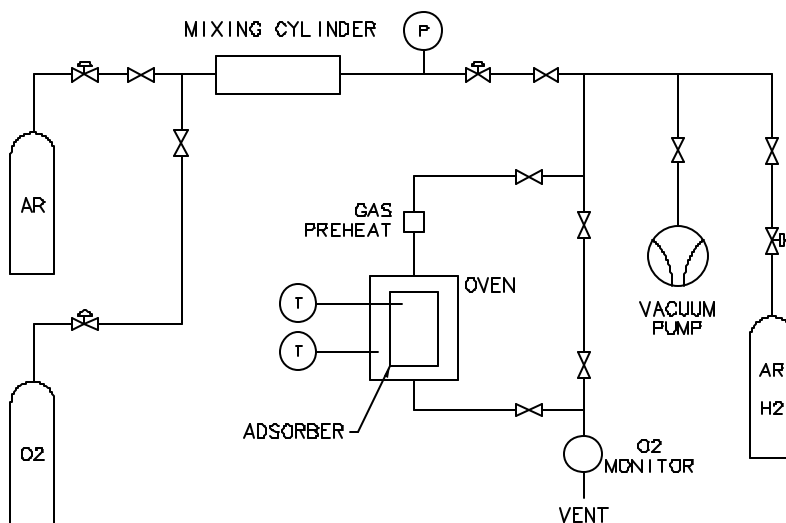

FIGURE 5. Simplified adsorber test setup.

TABLE 1. Regenerated Trigon adsorber test results.

	Adsorber Volume [mL]	Adsorber Mass [g]	Initial O2 Concentration [ppm]	O2 Mass Flow Rate [g/h]	Time to Saturation [min]	Total O2 Adsorbed [g]
Test 1	114	94.2	5500	0.346	94	0.54
Test 2	114	94.2	5400	0.340	70	0.40

The procedure for determining the purity achieved and total quantity of oxygen adsorbed is as follows. First, the mixing cylinder is filled with air to a known pressure, and thus a known quantity of oxygen. Then the mixing cylinder is filled to a much higher pressure with commercial-grade argon. The rest of the system is evacuated to insure there is no additional oxygen in the system. The argon-air mixture flows at atmospheric pressure from the mixing cylinder through a flow meter, then the adsorber, and finally to an oxygen analyzer. The mixture is allowed to flow through the adsorber until the oxygen monitor indicates that the adsorber is saturated and no longer removing oxygen. The total time until saturation, the flow rate, and the initial and final concentrations of oxygen in the argon-air mixture are recorded.

Two commercial adsorber pellets, Trigon and Messer, were tested and successfully regenerated. The two test results of regenerated Trigon adsorber pellets are presented in TABLE 1. For calculating the total amount of oxygen adsorbed it was assumed the entire oxygen concentration was removed. Our oxygen analyzer could only read down to 0.1 ppm and was consistently seen to bottom out during purification. Therefore, a regenerated sample was sent to ICARUS for further testing of its purifying capabilities. They determined that an impurity concentration of around 0.1 ppb oxygen equivalent could be attained with the regenerated adsorber.

With the knowledge about in-line regeneration from the adsorber test setup, the design of a liquid argon purification system could begin. The current simplified design of this liquid argon purification system is shown in FIGURE 1. Before any argon enters the system, all the surfaces are cleaned with diluted nitric acid, rinsed with de-mineralized water, and dried with dry air. The entire system is then evacuated to a high vacuum to insure that most air is removed. Commercial liquid argon is pumped through a molecular adsorber to remove any polar molecules such as water, which degrade the performance of oxygen adsorbers. From there the argon proceeds through an oxygen adsorber, and then enters the storage dewar.

The liquid argon may need to be cycled back and forth a couple of times between the storage dewar and the detector chamber to achieve the necessary purity levels. There are two main factors which gradually decrease the purity level over time. The first is the constant leaks of air through all the seals and joints in the system. The second factor is the outgassing of the inner walls, cables, and electronics which is expected to decrease over time. As a result, a constant purification process must be established in order to maintain the purity levels desired.

MONITORING PURITY

By using the innovative methods developed by ICARUS, the free electron lifetime can be measured and the corresponding purity of the liquid argon can be inferred. Moreover, the measurement of the electron lifetime is of direct interest, since electrons produced by

the ionization in the LAr must have drift times on the order of several milliseconds, without considerable attachment to electronegative impurities, for optimum physics results [1].

See FIGURE 1 for a simplified schematic of the purity monitor. The purity monitor composed of an anode and cathode disk separated by a drift distance, is immersed in the LAr. A photon light source outside of the vessel emits a short pulse of high energy light. The laser pulse is transmitted via fiber optic into the cryostat and is directed onto a photo-cathode located at the center of the cathode disk. Millions of electrons are released in all directions by way of the photo-electric effect. The photo-produced electrons begin to drift towards the anode along the applied electric field. A fine grid plane very near the cathode disk measures the initial charge (Q_C) of the total electrons extracted. A second grid is located at the opposite end of the drift distance, just before the anode disk, and measures the final charge (Q_A) of the remaining electrons. As the electrons drift from the cathode to the anode, some will come in contact with and attach themselves to electronegative impurities. As a result, there is a decrease in electron charge measured at the anode end of the drift distance. The time it takes for the electrons to travel from the cathode to the anode drift time (t_d) is dependent on the drift distance and applied electric field. Focusing rings are equally spaced along the drift distance to insure a uniform electric field and prevent false purity readings by insuring all electrons extracted stay within the drift volume and are lost only by attachment to electronegative impurities. The ratio of charges Q_A and Q_C is related to the drift time (t_d) and the electron lifetime (t), by the equation [7].

$$\frac{Q_A}{Q_C} = \exp\left(-\frac{t_d}{t}\right) \qquad (1)$$

Next the electron lifetime is related to impurity parameters.

$$t = \frac{1}{K \cdot N} \qquad (2)$$

Where, N is the impurity concentration and K is a function of the applied electric field. Since oxygen is the main source of electronegative impurities, parameters K and N are calculated in terms of oxygen properties and the overall impurity concentration is then referred to in terms of oxygen equivalent.
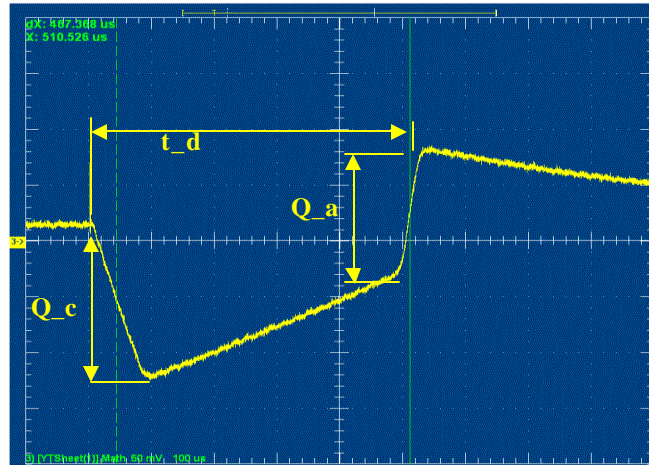

FIGURE 6. Oscilloscope waveform (charge versus time) produced in collaboration with ICARUS.

An oscilloscope waveform from a purity monitor readout is presented in FIGURE 6. The waveform was produced in collaboration with ICARUS, using an oxygen adsorber regenerated by FLARE. From FIGURE 6 with Q_C at 136 mV, Q_A at 112 mV, and t_d at 467 ms, applying equation (1) yields an electron lifetime t, equal to 2.4 ms. Lifetimes of greater than two milliseconds measured with the purifier specifications used in the ICARUS test setup are very good and meet the 0.1 ppb oxygen equivalent requirement. Therefore, the regeneration process has been successful and marks a FLARE R&D milestone.

CONCLUSION

The nearly completed experimental setup will be used to perform a series of tests to better understand the effects different variables have on the purification process. Of primary interest is the effect various materials, filling without evacuation and outgassing of warm surfaces, has on the argon purity and purification rate. Future experiments will extrapolate from the data approximations for speed of purification during initial fills, recirculation rates for maintaining purity, and dimensioning of purifiers. In addition, the performance (rate of saturation and level of purity achievable) for various industrial adsorbers such as Oxysorb, Trigon, and BASF will be tested.

ACKNOWLEDGMENTS

REFERENCES

1. Bartoszek, L. et al., "Letter of Intent," Fermilab Liquid ARgon Experiments, August 23, 2004.
2. Amerio, S. et al., "Design, construction and tests of the ICARUS T600 detector," 2004, Accepted for publication by Nucl. Instr. Meth. A.
3. Brentari, E. G. and Giarratano, P. J., *Boiling Heat Transfer for Oxygen, Nitrogen, Hydrogen, and Helium,* U. S. National Bureau of Standards, Boulder, Colorado, 1965, Technical Note 317.
4. Incropera, F. P. and DeWitt D. P., *Fundamentals of Heat and Mass Transfer*, John Wiley & Sons , New York, 2002, chapter 10.
5. Childs, G. E., Ericks, L. J. and Powell, R. L., *Thermal Conductivity of Solids At Room Temperature and Below,* National Bureau of Standards, Boulder, Colorado, 1973, Monograph 131, pp 212.
6. Benetti, P. et al., "A Three-ton Liquid Argon Time Projection Chamber," Nucl. Instr. Meth. A, 2000, pp. 395-412.
7. Arneodo F., "Electron Lifetime and Purity Monitor for the ICARUS T600 Detector," 2004, ICARUS-TM/02-14.